The Urey—Craig Groups of Chondrites
and Their States of Oxidation

Hans E. Suess

Department of Chemistry

University of California San Diego

La Jolla, California

Dedicated to
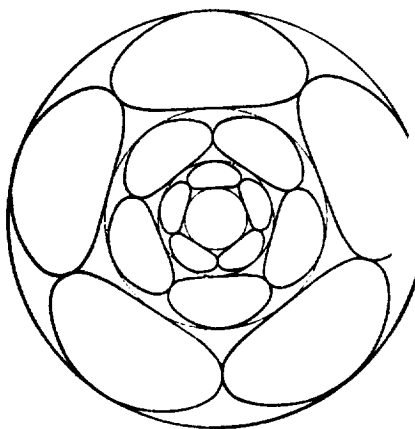
Harold C. Urey

on his seventieth birthday

1963

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Department of Chemistry
University of California, San Diego
La Jolla, California

Abstract

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Precise measurements by Keil and Fredriksson of the FeO content
of coexisting olivine and rhombic pyroxene in chondrites belonging
to the same Urey-Craig group show surprisingly small variations. The
FeO content of the silicates cannot be used as a measure for differ-
ences of the degree of oxidation of chondrites belonging to the same
group. No correlation between the FeO content of the silicates and
the metal content of the respective meteorite is apparent. Urey and
Craig's classification of the chondrites into two groups is more fun-
damental than a difference in the average iron content and the average
state of oxidation. It appears that the two groups contain some un-
altered material derived from two different homogeneous parent reser-
voirs.

Some aspects of the problem of the origin of chondrules are
discussed. It is suspected that the absence of a planet in the region
of the asteroids, and the constant FeO content of the silicate miner-
als in the Urey-Craig groups, have a common cause in the marginal
vapor pressure of water at the distance of the asteroids from the sun.
In this area the formation of large solid bodies may have taken place
in two steps: In the first step, water and other hydrides were solid.
Together with metal and silicate grains these solids accumulated into
relatively large, perhaps lunar sized planetesimals with relatively
high center temperatures. The combined heats of accumulation and of
radiation from the sun led to re-evaporation of the hydrides and to the
re-dissolving of these objects. Thereafter, separation of gases from
metal and silicate particles occurred. In a second step, metal and
silicates accumulated into objects of the composition of meteorites.
On the basis of such a complex picture, the existence of the various
types of meteorites and their properties can be explained by several
plausible ad hoc hypotheses.

Introduction

Our knowledge of the cosmic abundances of the non-volatile elements is primarily derived from the average composision of stony meteorites. Unsöld (1948), Brown (1949), and others have shown that this composition corresponds closely to the abundance values of the elements obtained from spectral analyses of the sun and of stars. In 1947, the author showed that the values derived by Goldschmidt (1938) were connected, though in a rather complicated way, with the nuclear properties of the individual isotopes of each element. From this it was concluded that cosmochemical separation processes had not substantially obscured the primeval abundance ratio of the non-volatile elements in meteoritic matter (Suess, 1947).

Yet the similarity between the average composition of meteorites and the content in the sun and in stars of non-volatile elements has its limitations. Increasing precision of observational data for meteorites and for the sun and stars indicates that the solar and the meteoritic abundances, though in general of the same order of magnitude, show marked deviations. Furthermore, abundance data for a number of elements do not seem to fit into a plausible picture derived from the abundance data of the other elements of the respective range of the periodic table. It has become obvious now, that chondritic meteorites do not reflect the abundances as precisely as was previously thought (Urey, 1952; Suess and Urey, 1956; Urey and Murthy, 1963).

The assumption that chondrites were representative of the non-volatile part of solar matter was based on the following argument: The primeval ratio of metal to silicate was not changed appreciably by the processes that led to the formation of meteorites; therefore, it appeared extremely unlikely that elements occurring in the same meteoritic phase and showing similar chemical properties should have been fractionated to a large degree by these processes. This argument, however, cannot be considered conclusive any longer. Urey (1961) has frequently pointed out that the four terrestrial planets cannot all have the same iron content, because their uncompressed density differs considerably. Urey states that Mercury, for example, has a density of 5.59, whereas the density of Mars is 4.22. Secondly, the discovery by Urey and Craig (1953) of the existence of two groups of chondrites, differing in their total content of iron, was evidence of a separation of iron from silicate during the formation of chondrites. Thirdly, more recent astronomical investigations of the solar abundance of iron tend to give considerably lower iron values than those estimated from the chemical analysis of meteorites. While the meteoritic values give an atomic Fe/Si ratio of 0.6 to 0.85, the solar abundance ratio is now being estimated to be below 0.5. Aller (1961) gives the Fe/Si ratio in the sun as low as 0.15. The importance of this discrepancy for any theory of the formation of the solar system has been emphasized by Urey (1960).

Whereas our knowledge of the primordial abundances of the elements was limited previously by the lack of reliable analytical date, we are now

arriving at a stage where further improvement cannot be achieved without some knowledge of the mechanism by which the meteorites formed and of the cosmochemical processes that were important during their formation. Urey (1960) has often expressed the opinion that the meteorites cannot have formed in a relatively simple manner from a primeval planetary nebula, but that they show signs of a rather complex and involved mechanical and chemical history. The basis for this conclusion is unusual because it is not a complex variety of chemical and petrological properties that indicates such a history. It is the unique monotony, the remarkably small variability in many properties and in bulk composition of meteoritic material, as compared with terrestrial rocks, that makes this conclusion necessary. An example illustrating this is discussed in this paper.

Oxidized iron in ortho-and metasilicate

Next to oxygen and silicon, magnesium and iron are the most abundant elements in meteorites. The meteoritic content of all other elements amounts to less than 10 per cent of the total number of atoms. Iron is present as metal, as sulfide, and in the oxidized state contained in the silicates. Prior's law (1920) states that the sum of the iron in the three different chemical forms is nearly the same for all chondrites. Urey and Craig (1953) have shown, however, that this is only partly true. They found two groups of chondrites, the so-called low and high iron group chondrites, for which the total iron content was markedly different. The average values for total iron were 22.33 and 28.58 per cent, by weight.

Figure 1 illustrates this: Constant total iron content corresponds to diagonal lines in the diagram, with a slope of about 45°, the deviation from this angle being due to the difference in oxygen content. The two lines in the diagram correspond to the two values for the total iron; i.e., the sum of oxidized plus non-oxidized iron. These values were determined as average for the two groups of chondrites by Urey and Craig from a number of carefully selected chemical analyses in the literature. Indicated in the figure also are individual values as given by Mason (1962) which include more recent analytical results. The new values confirm the conclusions of Urey and Craig, but tend to suggest somewhat lower values for the average iron content.

As can be seen from Figure 1, the low iron group of Urey and Craig is identical with the olivine-hyperstene chondrites of Mason (1962). The olivine-bronzite chondrites and the olivine-pigeonite chondrites of Mason belong to the Urey-Craig high iron group chondrites. The iron sulfide content is nearly the same for all these three groups.

Magnesium and oxidized iron occur in chondrules as ortho- and meta-silicates, in the form of two minerals which are called olivine and rhombic pyroxene, respectively. The relative amounts of the two minerals are stoich-iometrically determined by the ratio of the amounts of Si to Mg to Fe present in the particular chondrule, according to the equations given below. The Si balanced by other metallic elements, in particular Al, Ca, and Na (about 5 to 10 per cent) has to be subtracted from the total Si.
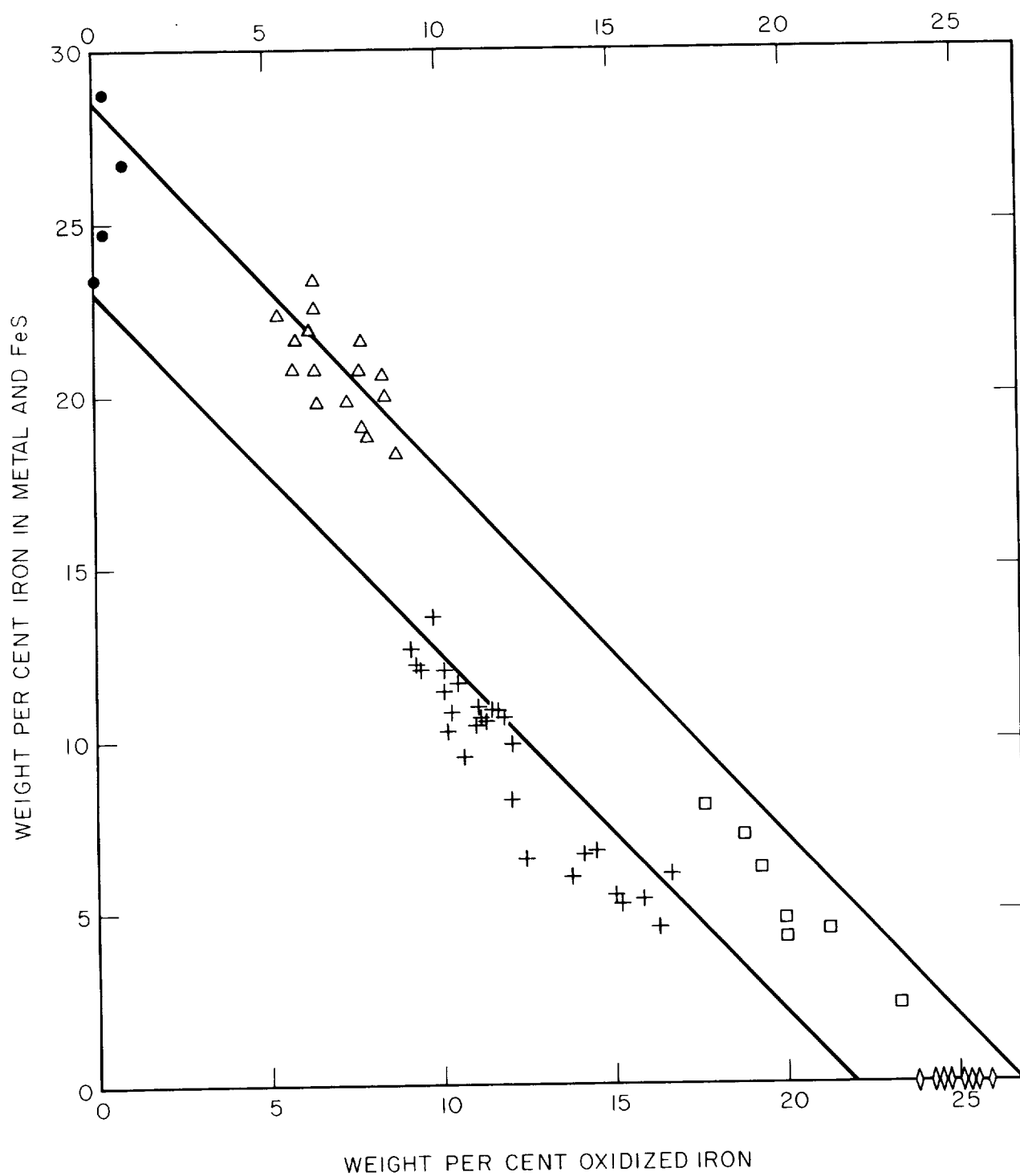
Fig. 1. Silicate phase iron plotted against metal phase iron and
sulfide, according to Urey and Craig. Individual points are taken
from Mason (1962).

Using the following notations: $N(MgO)$, $N(FeO)$, $N(SiO_2)$ as the number

of moles of MgO, FeO, SiO_2 per unit of weight present; $M(MgO)$, etc., and

$O(MgO)$,,etc., as the corresponding number of moles present in the meta-and

ortho-silicate, respectively, so that $N(Fe) = O(FeO) + M(FeO)$, etc., then

one obtains for the number of moles of ortho-and of metasilicate per unit

of weight $O(total)$ and $M(total)$:

$$O(total) = N(MgO) + N(FeO) - N(SiO_2)$$
$$M(total) = 2N(SiO_2) - N(MgO) - N(FeO)$$

Si and Mg are present in almost equal amounts and, therefore, the mole

content of oxidized iron $N(FeO)$ should be approximately equal to that of

the ortho-silicate $O(total)$,(Latimer, 1950). The equilibrium situation is

described by the reaction

$$MgSiO_3 + Fe\ (met) + H_2O = MgFeSiO_4 + H_2$$
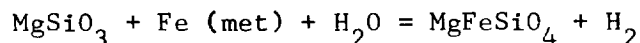
because metallic iron is always present in chondrites. However, under

equilibrium conditions, both olivine and pyroxene contain iron. The distri-

bution of the iron between the two minerals can be given as a Nernst co-

efficient α, which is approximately independent of the amount of oxidized

iron $N(FeO)$ in the system and is defined as:

$$\alpha = \frac{O(FeO)\ M(total)}{O(total)\ M(FeO)}$$

The measurements by Keil and Fredriksson (1963) give an average value

for the α measured for about 70 chondrites

$$\alpha = 1.13$$

with deviations of less than 10 per cent. A detailed discussion of the

Fe distribution in associated terrestrial minerals was published by

Bartholome (1962).

From the relationships given above, the iron content of the ortho-

and meta-silicate as a function of the oxidized iron in the system can be

calculated. The iron content of the olivine, for example, is found to be:

$$\frac{O(Fe)}{O(total)} = \frac{N(FeO)}{(2 - \alpha)N(SiO_2)-(1-\alpha)N(MgO)-(1-\alpha)N(FeO)} \tag{1}$$

If α is not very different from one, the iron content of the olivine,

as well as that of the pyroxene, will be proportional to the total oxidized

iron in the system.

This result is important because it shows that the amount of oxidized

iron, both in the olivine and in the pyroxene, should be a measure for the

total oxidized iron present and, therefore, should be a measure for the

degree of oxidation of the meteorite as a whole.

Oxidized iron in the low and high iron group chondrites

Work on the Bjurböle meteorite, as well as other investigations by

Fredriksson (1962), show that the FeO content of the silicates of a chon-

drite is frequently the same in all its chondrules. Optical measurements

of the iron content of the silicates in different chondrules had already

shown a relatively small variability for the individual meteorites

(Ringwood, 1961; Mason, 1962). Fredriksson's measurements with the more

precise X-ray microprobe have now confirmed that the iron content is indeed

surprisingly constant. The author, therefore, expected that the FeO,

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content of the minerals in the chondrules could be used as a good measure

for the state of oxidation of a chondrite. According to Prior's law, the

content of oxidized iron of the silicates should vary with the metal con-

tent in such a way that the sum of oxidized, metallic, and sulfidic iron

should be constant. The author, therefore, suggested measurements of the

FeO content of co-existing pyroxene and olivine in chondrules of a large

number of chondrites of both Urey-Craig groups. The measurements were

carried out by Dr. K. Keil, who enjoyed the advice and help of

Dr. K. Fredriksson during the work. For different reasons, Dr. Fredriksson

had planned independently similar investigations. The measurements were

carried out by means of an ARL Electron Microprobe X-ray Analyser in the

laboratory of Dr. G. Arrhenius. The elements Mg, Fe, and Ca were measured

simultaneously. A complete list of the experimental results is being pub-

lished by Keil and Fredriksson, (1963).

Figure 2 shows histograms of results of measurements by Keil and

Fredriksson (1963) and of data given by Urey and Craig (1953). One sees

that the spread in the FeO data is much smaller than that in the data ob-

tained from bulk chemical analyses. The two FeO peaks in the histograms

for olivine and for pyroxene correspond, as expected, to the two Urey-Craig

groups of metal content. As was mentioned before, the ratio of the FeO

content of olivine to that of pyroxene in the same meteorite is relatively

constant. The average value for this ratio α is 1.15 for L-group and 1.11
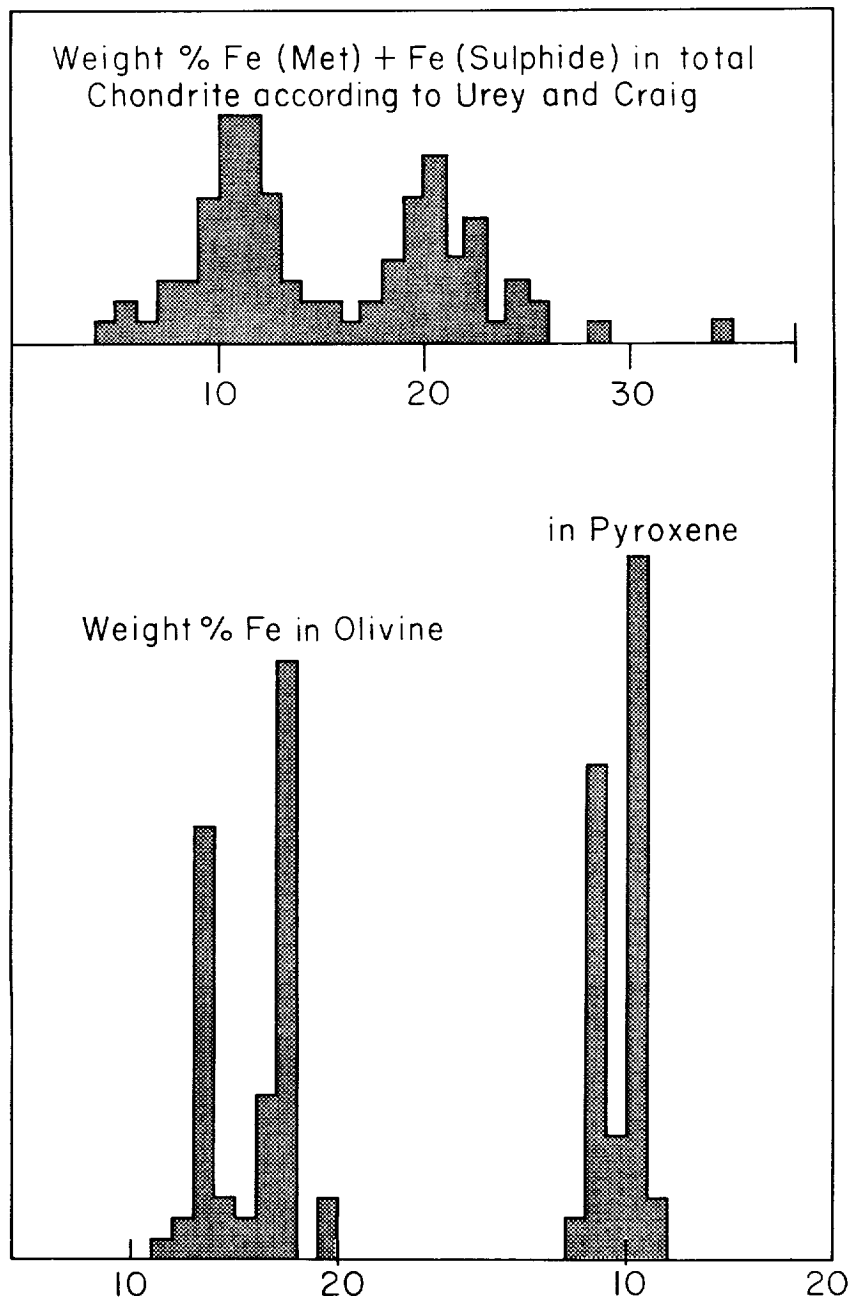
for H-group chondrites.

Fig. 2. Upper part: Histogram of metal content of chondrites, according to Urey and Craig (1963). The two histograms of the FeO content of olivine and pyroxene in the lower part of the figure represent data obtained by Keil and Fredriksson (1963), using electron micro spectroscopy X-ray.

Figure 3 shows a Urey-Craig diagram, showing the same diagonal lines

for low and high iron content as Figure 1. Individual values, however,

are shown only for meteorites for which Keil and Fredriksson measured the

FeO in the olivine and in the pyroxene. The measured FeO content of the

olivine and that of the pyroxene is shown to the right and to the left of

the respective point for the bulk chemical analysis. The points for each

meteorite are connected by a horizontal bar. In order to make the results

more comparable with each other, the iron content of the silicates, as

determined by the electron microprobe, is shown in Figure 3 on a 20 per

cent smaller scale than the data for the bulk analyses of the meteorites;

the difference corresponds approximately to the amount of the other mater-

ial, such as metal, sulfide, and other silicates in the meteorite. Where

no bulk content of oxidized Fe is indicated, the content of Fe in metal

and sulfide was taken from the phase integration data by Keil (1962).

If Prior's law were valid and, also, if the FeO content of the silicate

minerals were proportional to the total oxidized iron in the particular

meteorite, as required by equation (1), then the values for FeO content

of the olivine and of the pyroxene should lie on diagonal lines approxi-

mately parallel to those shown in Figure 3. The figure shows that the

results of the X-ray microprobe investigations of the silicates are by

no means in agreement with what is expected from Prior's law and from a

constancy of total iron for each group of chondrites. While the content

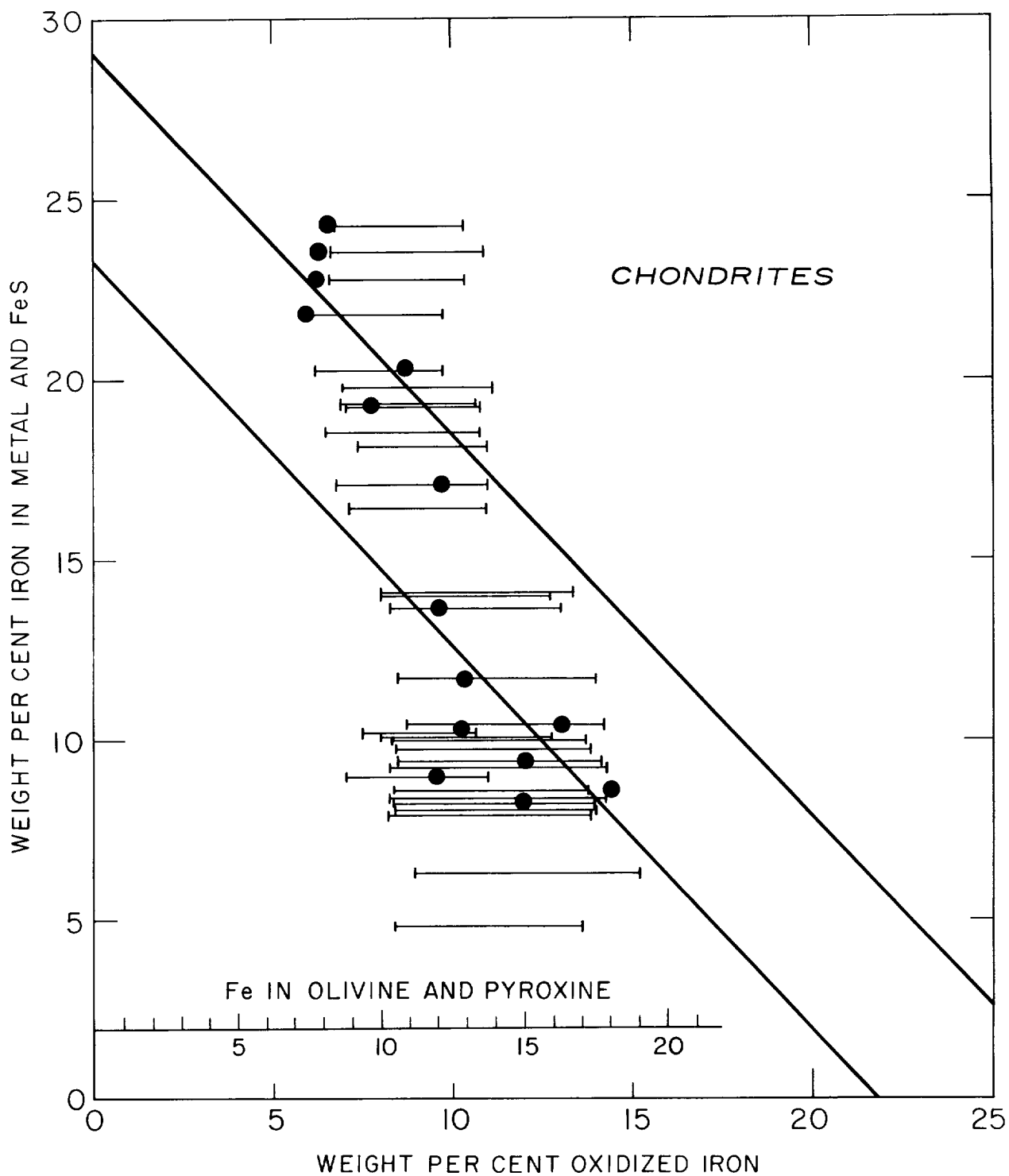of metallic plus sulfidic iron in the investigated meteorites varies by

Fig. 3. Urey-Craig diagram of silicate phase and metal phase iron.
Solid circles refer to chemically determined oxidized iron. The end
of the horizontal bars are values for the FeO content of olivine
(right) and pyroxene (left). These values are plotted on a 20 per
cent smaller scale for easier comparison. If Prior's rule were valid,
the end points of the horizontal bars would lie parallel to the diag-
onal lines.

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more than a factor of two, that of oxidized iron in olivine and pyroxene of the same meteorites shows fluctuations of less than 20 per cent. The fluctuations have no obvious correlation with the metal content.

Although there are definite indications from bulk chemical analyses supporting the validity of Prior's rule within each group of chondrites (Wiik, 1956), the content of oxidized iron of individual minerals so far investigated does not seem to agree with this rule. The more qualitative statement originally proposed by Prior (1920) may have some validity if referring to all types of chondrites. Perhaps this statement should be expressed in the following way: The content of metallic and of oxidized iron of chondrites shows larger variations than the total iron content.

In contrast to the findings described above, investigations of several carbonaceous and pigeonite chondrites gave completely different results. In chondrules from these meteorites, silicates with an irregularly varying FeO content were found. It appears that much further work is necessary before systematic trends in the composition of these meteorites can be resolved.

Fredriksson's paradox and the origin of chondrules

The careful and exhaustive investigation of the Bjurböle meteorite by Fredriksson (1962), using microprobe techniques, gave within narrow limits the same iron content for all the olivine in the meteorite. Also, the pyroxene crystals showed a constant Fe content. Fredriksson's observation is remarkable because, in general, cooling of a melt produces

olivine and pyroxene crystals with a varying iron concentration (Bowen and Schairer, 1935). According to Fredriksson (1962), rapid cooling of a melt and recrystallization in the solid phase will lead to a constant iron content in olivine. Complete thermodynamic equilibrium will, of course, also give a defined, constant iron content in both minerals. In any case, various observations indicate that the minerals in many chondrules crystallized from homogeneously molten droplets. One would expect that the melt from which the chondrules formed had a constant composition; this, however, cannot have been the case. According to Fredriksson (1962), the ratio of the amounts of ortho-and metasilicate varies over a wide range from chondrule to chondrule. In extreme cases, it appeared that a chondrule consisted almost entirely of one mineral only, either olivine or pyroxene. The FeO content of the two silicates in the chondrules,

however, rarely varies by more than 10 per cent. From this it must be concluded that the Mg/Si ratio in the different chondrules differs by more than a factor of two. The fact that the Mg/Si ratio appears to vary from chondrule to chondrule in one and the same meteorite, while the FeO content of the silicates remains essentially constant, may be called Fredriksson's paradox. Bulk chemical analyses of different chondrites give exactly the opposite results.

The observations on the Bjurböle meteorite by Fredriksson can perhaps be explained in one of the two following ways:

1. The individual molten droplets that resulted in the formation of

chondrules formed from aggregates of varying amounts of ortho-and meta-silicates. In other words, the chondrules were formed in a second high temperature stage which melted mineral particles that had formed earlier and were present as dust and sand grains.

2. The chondrules were in constant thermodynamic equilibrium with each other through evaporation and condensation from a gas phase of sufficiently high pressure. In this case, droplets containing varying amounts of mineral crystals can form as a consequence of random effects from surface tensions, supercooling, and crystallization rates.

In any case, Fredriksson's observations are a further indication for a rather complicated history of meteoritic matter, as often postulated by Urey.

At first sight the concept of chondrules forming as the first condensation products directly from the gas phase, appears very attractive. At temperatures above the melting point of iron and silicates all the Fe in equilibrium with a gas of primordial composition will be reduced to metal. Chondrules contained in enstatite chondrites could conceivably have formed directly by condensation from a gas of primordial composition and sufficiently high pressure. This is shown in Figure 4. The partial pressures of various compounds (Urey, 1950) are plotted against temperature in a system containing the elements in primordial ratios. A hydrogen pressure of 350 atmospheres is chosen as an example. With such a high pressure, liquid condensation products of the composition of enstatite chondrites
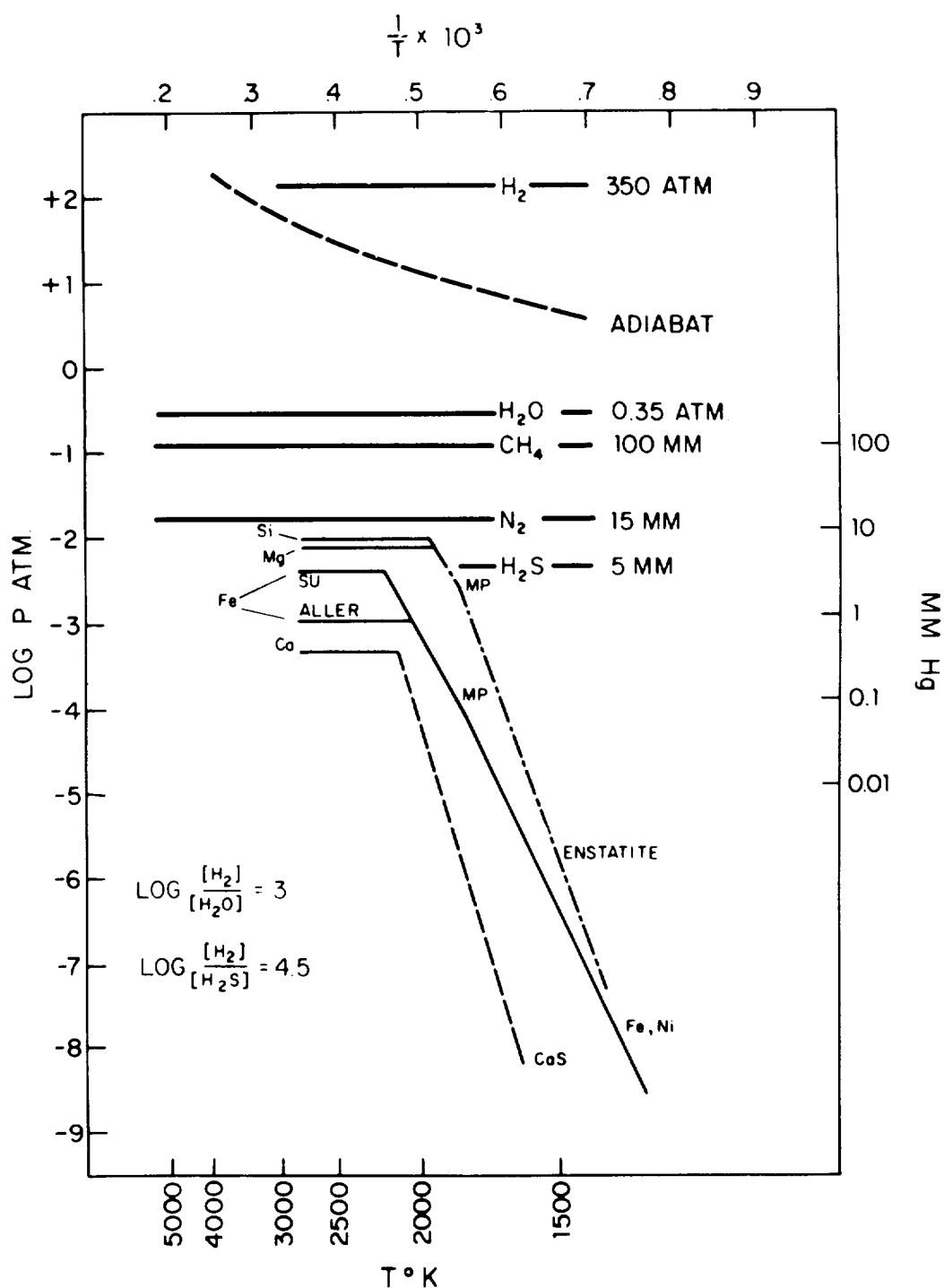
Fig. 4. Composition of gas phase in $\log_{10}$ of atmospheres plotted against reciprocal abs. temperature for gas of primeval composition and a partial H_2 pressure of 350 atm. Melting points MP of the condensed Nickeliron and Enstatite are indicated in the figure.

will form. At lower pressures, however, condensation will not occur above the triple point of iron and silicates, and the material will condense into solid crystals and not into droplets that could have solidified into chondrules.

In order to obtain chondrules that contain oxidized iron through condensation from a gas phase, the ratio H_2/H_2O has to be of the order of one instead of one thousand. In 1949 the author suggested, in connection with a theory for the formation of the earth's core proposed by Eucken (1948), that the lightest gases, mainly hydrogen and helium, escaped continuously from the gas sphere in which condensation of chondrules took place. In this way, the H_2/H_2O ratio decreased steadily during the course of condensation (Suess, 1949). Enstatite chondrites, according to this model, represent products of early condensation; Ca-rich achondrites, terrestrial surface material, and tektites, products of late (rest) condensation. However, Urey (1952) pointed out to the writer the many serious difficulties of this picture. In particular, he showed that thermodynamic data better than those available to the writer at the time when the suggestion was made, would indicate that condensation of many elements would not follow the sequence represented by the composition of the materials.

A further difficulty is the necessary gas phase separation of H_2 from H_2O by three orders of magniture. The experimentally observed He - Ne separation in meteorites should be of comparable order.

Empirical data, and possible explanations of them, were discussed in detail by Signer and Suess (1963). Fredriksson's paradox, as well as the nearly constant FeO content of the silicates in the Urey-Craig group chondrites, show that the meteorites cannot have formed in such a simple manner as was suggested in 1949.

It was recently suggested by Wood (1962) that the chondrules obtained their content of oxidized iron through metamorphic processes in the solid phase. Other authors (for example, Mason, 1962) have expressed the opinion that chondrules might have obtained not only their final chemical composition, but also their geometric form through diffusion processes in the solid state. The observations by Fredriksson and by Keil and Fredriksson, in particular the remarkable constancy of the FeO content of the minerals of each group, as well as other evidence, speaks against such assumptions. The existence of the two Urey-Craig groups of chondrites, each with an almost constant FeO content of their main minerals, makes it necessary to assume that each group of chondrites formed in homogeneous environment with well-defined redoxy conditions. An atmosphere of appropriate chemical composition constitutes such an environment. Separation of hydrogen from water at an early stage, followed by reheating to high temperatures above the melting point of the silicates, may have provided the necessary conditions.

For the two groups, the olivine-bronzite and the olivine-hypersthene chondrites, conditions must have been somewhat different, so that it is tempting to assume that each group came from a different parent body.

It is also possible that they came from two groups of parent bodies, each of which formed at a slightly different distance from the sun. Partial separation of metal from silicate must have preceded formation of the bodies and it must have occurred to a slightly different degree for the two cases.

Parent bodies of meteorites

It is in general agreed that meteorites, or at least the majority of them, come from the asteroidal belt. They are presumably produced continuously through collisions of asteroidal bodies, or their fragments, with each other (Arnold, 1963). The asteroids occupy the space between the inner, terrestrial, and the outer, jovian planets. Their orbits cover an area of the solar system where obviously, according to the law of Titius and Bode, a planet is missing. The large masses and low densities of the outer planets, compared to masses and densities of the inner ones, are caused by a high content of water, ammonia, and methane (Wildt, 1938, Brown, 1950). Urey (1952) has shown why H_2O, NH_3, and CH_4 were retained by the planets that formed beyond the asteroidal belt and why these hydrides were lost by those that formed nearer the sun. It is essentially the vapor pressure of water that caused the difference. As was pointed out by Urey (1952), the vapor pressure of water as a function of the distance from the sun with its present luminosity is negligibly small beyond the asteroidal belt. At lesser distance, however, it is such that water will be gaseous in the disc of primeval solar material from which the planets formed.

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It seems most plausible to assume that the formation, in a transitional area of the solar system, of a number of small bodies rather than a single planet, was also a consequence of the magnitude of the vapor pressure of water at this particular distance from the sun.

Astronomical considerations lead to the conclusion that at the time of formation of the solar system a very effective process must have led to a continuous, almost quantitative separation of gases from condensed material (Kuiper, 1952, Signer and Suess, 1963). Presumably the early planetary nebula contained water and hydrides in the form of frozen particles (Fowler and Hoyle, 1962), from which hydrogen, helium, and other highly volatile gases were rapidly separated by this process. In the range of the inner planets, the small, ice-containing particles accumulated into larger bodies (dirty snowballs), until they had become large enough so that direct radiation from the sun could reach them. This was the case when the material had accumulated into objects of the order of kilometers or tens of kilometers in diameter. Thereafter, radiation from the sun led to a rise in temperature and to evaporation of the ice. Following this, the water vapor and other gases were separated from the solids, which finally accumulated into the inner planets and their satellites.

Quantitative estimates of the conditions that prevailed temporarily and that were determined by rates of accumulation of solids, of reevaporation of ices, and of gas-solid separation are extremely difficult, but it appears plausible that the maximum size that was reached by the ice-containing

bodies increased with increasing distance from the sun. Beyond the aster-
oidal belt, radiation from the sun was insufficient to cause ice and
hydrides to evaporate (Urey, 1952) and the material accumulated into giant
planets.

Intermediate conditions that prevailed in the range of the asteroidal
belt were obviously not favorable for the formation of either an inner,
terrestrial, or a giant, jovian planet. One may assume that radiation from
the sun was insufficient to cause evaporation of ice, but in combination
with the heat of accumulation it may have led to conditions under which the
primary planetesimals dissolved. This may not have happened until lunar
sizes were reached. Temperatures and pressures within such protoasteroids
have been calculated by Bainbridge (1962). The calculations show that tem-
peratures exceeding the melting points of iron and of silicates were reached
in the interior of contracting lunar-size gas spheres. The interior of such
objects may have provided the proper environment and the necessary homogen-
eous redoxy conditions for the formation of the material contained in
chondrites.

On the basis of a different argument, Urey (1956, 1957) had also con-
cluded that the meteorites developed in two successive generations of bodies
The argument presented above cannot be directly coordinated with Urey's
original ideas without some modifications. In any case, the assumption of
two generations of meteoritic parent bodies — grandparent bodies of lunar
size, containing a large fraction of water and with internal temperatures

up to $2000^{\circ}K$, and parent bodies consisting of chondritic and other material similar to that of the inner planets — takes adequate care of the requirement of a complicated history of meteoritic matter. It leaves ample room for ad hoc hypotheses for explanations of the existence of the various types of meteorites, of the special and general anomaly of Xenon, of the deficiency of sulfur and of many other elements in chondrites, and of other facts. It limits, however, the degree to which we can expect similarities in the chemical composition of chondrites and of planets. Because of the more complicated history of the chondrites, their state of oxidation, their loss of certain elements, and their metal-silicate ratio may be considerably different from that of the inner planets.

Acknowledgement

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